Investigation of Effects of the Electronic Stopping Power in Molecular Dynamics Simulation of Radiation Damage in bcc-W

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1. Introduction

Materials of nuclear fission and fusion reactors are used in harsh environments such as high neutron flux, ion flux, and heat flux. For fusion devices, tungsten (W) has been widely studied as plasma-facing material because of its attractive properties, such as high melting point and high thermal conductivity [1].

In the radiation environment, collisions of incident ions and neutrons with atoms in solid materials produce energetic atoms called primary knock-on atoms (PKA), which leave their lattice sites with excess kinetic energy [2]. PKAs induce complex collision cascades and produce radiation defects, such as vacancies and self-interstitial atoms (SIAs), and their clusters, such as voids and dislocation loops. As a result, some material properties are deteriorated as radiation effects. Since radiation effects are directly linked to the safety and economy of the nuclear power system [3], it is important to understand how radiation damages occur and how they evolve in materials.

To study radiation damage by computer simulations, molecular dynamics (MD) simulations have been widely used [4]. However, in previous studies, there is inconsistency in the treatment of the electronic stopping power (ESP) [5][6], which is energy loss due to electronic excitation processes. Specifically, most studies ignored ESP, while some showed its significant effects on the number of radiation defects and morphology. In this study, we investigated the effects of ESP in MD recoil simulations of W for various PKA conditions and simulation settings.

2. Method

Collision cascade simulation was performed using classical MD code LAMMPS [7]. The potential model used in this study is an embedded-atom method (EAM) potential [8] parameterized by Derlet et al. [9] and modified by Bjorkas et al. [10].

After equilibration at 30 K and 1 bar with an NPT ensemble, a collision cascade was initiated by introducing excess kinetic energy to one W atom, which becomes the PKA, and was simulated under the NEV ensemble. To consider energy losses by electronic interaction during collisions, the ESP was implemented by applying a friction force to each atom with equation (1).

$$\vec{F}_i = \vec{F}_i^0 - \frac{\vec{v}_i}{|\vec{v}_i|} \times S_e \quad (1)$$

Here, $\vec{F}_i$ and $\vec{F}_i^0$ represent the resulting and original forces applied to the i-th atom, respectively, $\vec{v}_i$ is its velocity, and S_e is the ESP. The ESP values were obtained as a function of its velocity from the SRIM code, which is based on theoretical analysis and experimental data [11]. In addition, the cut-off energy, the lowest energy at which ESP is applied, was used to avoid quenching all the thermal modes. We adopted the cut-off energy as 10 eV as recommended in a previous study [6].

We performed collision cascade simulations with 28 conditions: 14 PKA energies (1, 2, 4, 6, 8, 10, 20, 30, 35, 40, 50, 75, 100, and 150 keV) and 2 PKA directions (<111> and <321>). For each condition, we prepared two cases where ESP was applied or not applied (No-ESP) to evaluate the effect of ESP. To consider the stochastic variance of irradiation damage, we collected 15 samples for each case. A simulation cell was prepared for each condition so that radiation defects do not cross the boundaries of the simulation cell. Each recoil simulation proceeded for 10~40 ps until defect structures converged. After the recoil simulation, generated defects were identified by the Wigner-Seitz defect analysis method [12].

3. Results

Figure 1 shows, for example, a defect evolution process induced by a PKA of 100 keV and direction <111> with the ESP. The blue and red dots indicate vacancies and SIAs, respectively. First, in section 3.1, the effect of the ESP on collision cascade dynamics, corresponding to the evolution from Figure 1(a) to (d), is analyzed. Next, the effect on the final defect structure, such as the number and geometry of radiation defects in Figure 1(d), is analyzed in section 3.2.

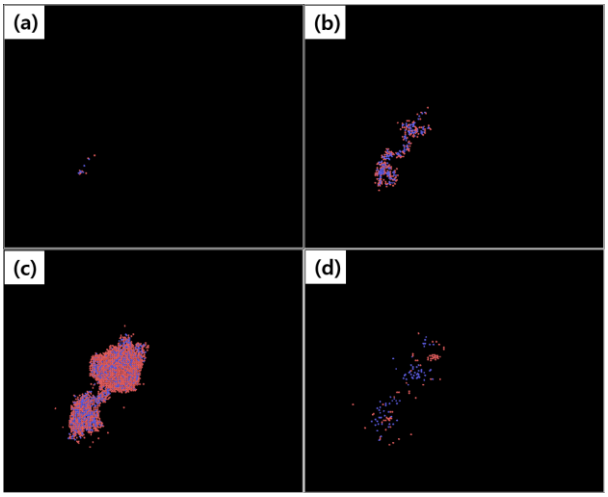

Figure 1. Defect structures generated by a PKA of 100 keV and <111> direction. (a) a recoil event starts, (b) a collision cascade occurs, (c) the defect number takes a maximum value, and (d) a stable defect structure is achieved at around 10~40 ps.

3.1. Collision cascade dynamics

To analyze the radiation damage dynamics, the time evolution in the number of Frenkel pairs obtained in each MD simulation was fitted with a Gaussian, and then averaged over 15 samples. Figure 2 presents the comparison for 8 simulation settings which are different in (i) PKA energy (10 keV or 100 keV), (ii) displacement direction (111 or 321), and (iii) with/without ESP. For the same PKA energy, the time to reach the peak point of Frenkel pairs was not affected by ESP and PKA direction. However, the peak number of Frenkel pairs was significantly overestimated when ESP was not considered: around 1.2 times for 10 keV and 1.65 times for 100 keV.

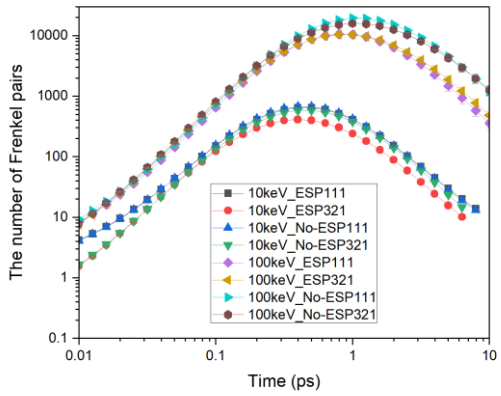

Figure 2. Time evolution in the number of Frenkel pairs generated during collision cascade simulations for several simulation settings.

3.2. Defect morphology after collision cascade

Figure 3 shows the number of surviving Frenkel pairs (N_f) as a function of PKA energy for each PKA direction with/without ESP. For both PKA directions, N_f increases with increasing PKA energy in a linear manner, approximately. However, at a certain point, which is denoted as transition energy hereafter, the slope changes and the number of surviving Frenkel pairs starts to increase rapidly, as observed in a previous study [5].

The transition energy was different between ESP and No-ESP simulations. The transition energy was calculated in two steps. First, a simple linear regression (SLR) was performed by adding data one by one from the lowest PKA energy and then finding the energy at which the fitting residual increased consecutively. Subsequently, SLR was performed for each side, and the intersection point of the two SLRs was regarded as the transition energy. The transition energy was almost the same in <111> and <321> for the No-ESP simulation, about 47 keV. However, with ESP, <111> has much lower transition energy (36.8 keV) than <321> (61.6 keV), as seen in Figure 3.

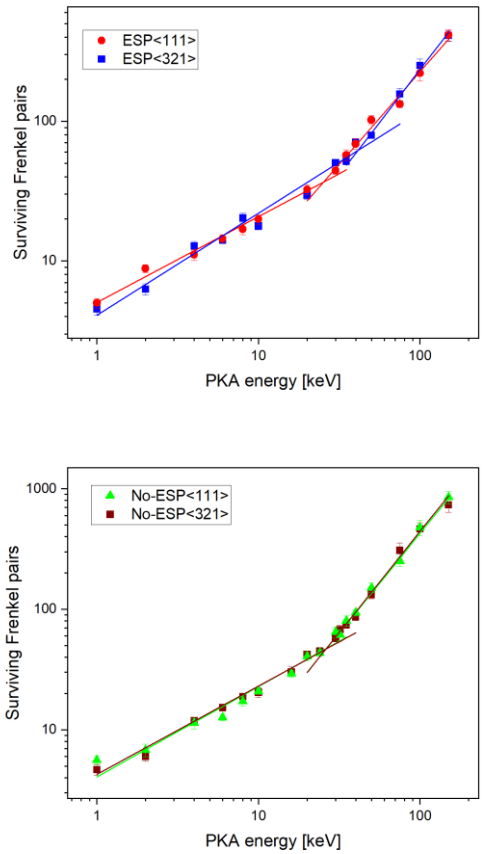

Figure 3. The number of surviving Frenkel pairs for ESP simulation (upper) and No-ESP simulation (lower). The lines indicate linear regression results.

To further analyze the defect structure morphology, the size of the largest SIA cluster (N_{max}) when the defect structure converged, e.g., Figure 1(d), was measured and compared with a previous study [6] in Table 1. The PKA energy was fixed as 150 keV in this comparison. The present MD simulation settings were almost identical to those in the previous work, except for equilibrating the bcc-W at 30 K in the present study and 0 K in the previous study [6]. No-ESP simulations always significantly overestimated N_f and N_{max} values, indicating the importance of ESP consideration. Although <100> case has the lowest N_f and the smallest N_{max}, the direction dependence is not large.

Table 1. Average number of Frenkel pairs N_f, the size of the largest SIA cluster N_{max}, and the number of simulation sample N_{sim}.

Direction	ESP	N_f	N_{max}	N_{sim}
<100>[6]	O	184±27.0	140	9
<110>[6]	O	202±31.8	233	9
<111>	O	207±20.2	170	15
<321>	O	205±18.0	191	15
<111>	X	425±48.8	365	15
<321>	X	367±46.7	278	15

3.3. Validation of empirical correction for ESP effect

In previous studies that did not consider ESP in collision cascade MD simulations, the effect of ESP was sometimes corrected afterward, assuming that the MD PKA energy is equivalent to the damage energy, which is the energy used in atomic processes to form defects. Under this assumption, the MD PKA energy in No-ESP simulation can be converted into the effective PKA energy as [Effective PKA energy] = [MD PKA energy] + [energy used in electronic processes]. The last term can be determined by a theoretical model, such as the NRT model [13]. By this empirical correction, for example, a No-ESP MD simulation with an MD PKA energy of 100 keV is considered equivalent to an ESP MD simulation with a PKA energy of 140 keV.

Figure 4 compares the number of surviving Frenkel pairs between ESP simulation and corrected No-ESP simulation. The x-axis is the PKA energy for ESP simulation and the effective PKA energy for No-ESP simulation. Below 75 keV, the differences between the two simulations are within statistic errors. However, when the PKA energy was above 100 keV, corrected No-ESP simulation clearly overestimated the number of surviving Frenkel pairs. This observation indicates that, although the empirical correction is sufficiently accurate at low PKA energies, ESP needs to be explicitly considered in collision cascades induced with high PKA energies, above 75 keV in the case of W.

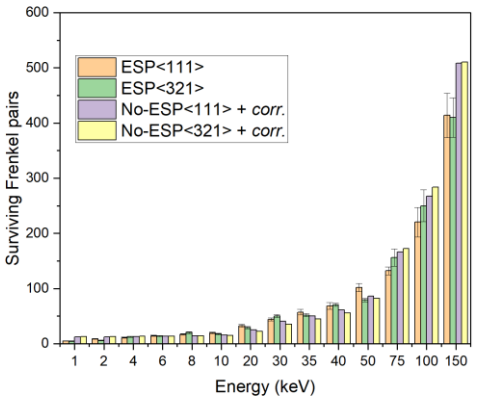

Figure 4. Comparison of the number of surviving Frenkel pairs as a function of PKA energy between ESP simulation results and corrected No-ESP simulation results.

4. Conclusion

In this study, we investigated the effects of ESP in recoil MD simulations. It was found that the ESP significantly changes not only the defect formation dynamics but also the number of survived Frenkel pairs. In addition, the empirical correction of ESP effects in No-ESP MD simulation does not work sufficiently at high PKA energies, above 75 keV in the case of W. These results indicate that the ESP needs to be properly considered in MD simulations when the recoil energy is high, such as radiation defects formed by fast neutron.

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